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14. ABSTRACT

This research explored new ways for diode laser fabrications. Focused was on the development of efficient organic light emitting materials and the fabrication of laser structures incorporating these materials. Very large optical gain was found due to stimulated emission in semiconducting conjugated polymers. Then we demonstrated that the huge light amplification can be used to build planar and ring lasers of very small sizes. We also investigated the optical properties of novel glass for amplifier and laser applications at communication wavelengths. We demonstrated light amplification at 1.3 micrometers and efficient room-temperature photo-luminescence in glasses doped with PbS quantum dots. Our efforts resulted in 18 publications and 13 contributions to international conferences including 4 invited talks.

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Summary

During this project, we explored new ways for diode laser fabrications. We focused on the development of efficient organic light emitting materials and the fabrication of laser structures incorporating these materials. We found very large optical gain due to stimulated emission in semiconducting conjugated polymers. We demonstrated that the huge light amplification can be used to build planar and ring lasers of very small sizes. We also investigated the optical properties of novel glass for amplifier and laser applications at communication wavelengths. We demonstrated light amplification at 1.3 micrometers and efficient room-temperature photo-luminescence in glasses doped with PbS quantum dots. Our efforts resulted in 18 publications and 13 contributions to international conferences including 4 invited talks.

A. Light Amplification in Conjugated Polymers and Organic Laser Diodes

Conjugated polymer semiconductors have attracted attention lately for device applications in areas that are classical domains of inorganic semiconductors such as electronic devices, e.g., diodes and field effect transistors, and photonic devices, e.g., holographic storage, light modulators, and light-emitting diodes. Structural flexibility, easy processing, polymer engineering through chemistry, and low cost make polymer devices competitive to devices based on inorganic semiconductors and their highly developed technology. The prospect of compact electrically pumped polymer lasers using their semiconductor properties is an exciting possibility.

We demonstrated that stimulated emission and very large optical gain can be achieved in semiconducting conjugated polymers. The gain covers a spectral range of about 50 nm in a region where the polymer is transparent and can be as high as 10^4 cm^{-1}. Utilizing the large light amplification in the polymer we obtained laser action for various planar and ring cavities of very

small sizes that are suitable for applications as diode lasers in integrated optics.

A1. Stimulated Emission and Optical Gain

To select suitable candidates for polymer laser diodes we studied study the nonlinear optical response and its dynamics after excitation with short optical pulses. Photoluminescence studies on thin films of our most promising candidate BEH:PPV (Poly(2,5-bis(2'-ethyl-hexyloxy)-1,4-phenylenevenylene) showed that above an average exciton density of $5x10^{18}$ cm^{-3} stimulated emission (SE) dominates the emission process. At this density the initially broad emission spectrum collapses into a sharp emission line and both emission intensity and quantum efficiency increase dramatically.

As a next step we performed direct measurements of optical gain spectra using femtosecond pump-probe techniques. In a broad spectral region (ca. 50 nm) starting just below the band gap energy huge gain coefficient of up to 10^4 cm^{-1} were measured in this material, by far the largest value reported for a semiconducting polymer. We found a cross-section for SE of $2x10^{16}$ cm^{-2} that indicates the possibility of considerable amplification even at densities below $5x10^{18}$ cm^{-3}. Time-resolved measurements showed that the initial gain decays rapidly on a 2 picosecond scale due to a reduction of the excited density by SE. For applications we can exploit the fact that about 10 % of the initial optical gain is still present after more than a nanosecond. Our studies revealed that the energy level scheme of BEH:PPV is similar to a four level system, known to be well suited for laser applications.

A2 Laser Action

The huge optical gain makes semiconducting conjugated polymers promising candidates as new laser materials. For laser applications, however, the polymer as the active material has to be combined with a structure that provides optical feedback. Laser action requires that the optical gain at least balances the total loss of the cavity structure. Since light amplification per unit length in semiconducting polymers is found to be very large small planar and ring laser configurations can provide sufficient optical feedback. This fact makes laser diodes utilizing solid-state organic materials particularly interesting for integrated optics applications. Although electrically pumped laser diodes are the final target, optical pumping is a suitable method to study the properties of different laser diode structures. We demonstrated laser action for various small resonator structures under optical excitation.

The most straightforward way to provide feedback is to design a cavity that uses two plane mirrors. To realize a high quality planar Fabry-Perot resonator dielectric mirrors are fabricated that

contain a stack of 21 alternating quarter-wave layers of low (SiO_2) and high (TiO_2) refractive index materials. These mirrors are characterized by low losses and extremely high reflectivities (>99 %) in the wavelength region of optical gain in the polymer. By spin-coating the polymer onto one of the dielectric mirrors and properly adjusting the second mirror we produced a low loss, high Q planar microcavity of about 9 μm optical thickness. In the presence of the cavity with BEH:PPV as the active material we observed laser emission for two longitudinal modes at 605 nm and 626 nm above a threshold of 50 $\mu J/cm^2$ excitation pulse energy. Above this threshold the laser emission increases linearly with excitation. The whole laser output is concentrated in an emission cone of smaller than 3^0 half-apex angle (i.e., it is diffraction limited); a striking evidence for spatial coherence. We also observed a high degree of polarization of better than 50:1 parallel to the exciting laser; further evidence for coherent laser emission. In contrast, neither the SE of the thin film nor the cavity emission below lasing threshold has a noticeable degree of polarization.

Another example for a planar resonator is a distributed feedback (DFB) laser structure fabricated by writing a surface relief grating of Λ=500 nm into a glass substrate using electron beam lithography and depositing the polymer (200 nm) on top of this grating. To get sufficient feedback by the grating and achieve polymer laser action the excitation light has been focused to a stripe of 3 mm length and 50 μm width. The laser emission of only 3.5 nm width is considerable narrower than the 8 nm wide SE band observed by exciting a part of the same film without a grating. For this structure we observed a lasing threshold of about 40 $\mu J/cm^2$ excitation pulse energy. However, it should be noted that this structure has not been optimized so far, and relatively large scattering losses as well as a dependence on the spot position due to a non-uniform grating have also been observed.

As an alternative to cavity configurations with plane mirrors and gratings, we studied two different ring resonator structures. The first microring is formed by the surface tension of a polymer droplet encircling an optical fiber of 16 to 120 μm diameter. This kind of structure forms a high quality cavity for whispering-gallery modes (WGM's). These modes propagate around the edge of the coated fiber with a strong optical confinement in the gain material. We demonstrated that the gain in a conjugated polymer is sufficient to achieve laser oscillations of several narrow and isolated WGM's. The lasing thresholds were in the 1 to 10 $\mu J/cm^2$ range and decreased with the fiber diameter. Between 300 and 10 K we found only a weak temperature dependence for the lasing threshold. Our results suggest that a significant threshold reduction can be achieved for low temperature operation if the quality factor of the ring cavity (at present on the order of 1000) is further improved.

We fabricated a second ring resonator laser structure based on a Si_3N_4 ring waveguide with a ring diameter of 35 μm. The waveguide was formed by a 25 nm high and 9 μm wide Si_3N_4 ridge

on top of a 275 nm Si_3N_4 layer that is deposited on SiO_2 substrate. The waveguide chip was afterwards covered with 50 nm of SiO_2, while an opening at the ring resonator position was provided. In the next step, a 100 nm thick polymer film has been spin-coated on the waveguide chip. Since the guided ring cavity mode reaches out into the polymer layer optical gain in the polymer leads to amplification of light that is emitted into one of the ring cavity modes. The ring resonator is designed to have a small coupling to a channel waveguide. We excited the active polymer material from the top with a pulsed laser and detected the output of the channel waveguide. Increasing the excitation we observed lasing of several ring resonator modes starting at 130 $\mu J/cm^2$ excitation pulse energy. Although this threshold was slightly larger than for the other cavities, this kind of structure is of particular interest since it can be easily integrated and the emitted light can be coupled from the waveguide into an optical fiber.

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